WHAT IS CLAIMED IS:

1. An article comprising:

a metallic substrate; and

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- a substantially single-phase coating disposed on said substrate, wherein said coating comprises nickel (Ni) and at least about 30 atomic percent aluminum (Al), wherein said coating further comprises a gradient in Al composition, said gradient extending from a first Al concentration level at an outer surface of said coating to a second Al concentration level at an interface between said substantially single-phase coating and said substrate;
- wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.
 - 2. The article of claim 1, wherein said coating further comprises at least one of chromium (Cr), zirconium (Zr), up to about 20 atomic percent cobalt (Co), and up to about 20 atomic percent iron (Fe).
- The article of claim 2, wherein said Cr is present at a concentration of up to about 15 atomic percent.
 - 4. The article of claim 3, wherein said Cr is present at a concentration in the range from about 4 atomic percent to about 12 atomic percent.
- 5. The article of claim 2, wherein said Zr is present at a concentration of up to about 20 2 atomic percent.
 - 6. The article of claim 5, wherein said Zr is present at a concentration in the range from about 0.2 atomic percent to about 0.8 atomic percent.

- 7. The article of claim 2, wherein said coating further comprises at least one element selected from the group consisting of hafnium (Hf), yttrium (Y), silicon (Si), titanium (Ti), lanthanum (La), cerium (Ce), and tantalum (Ta).
- 8. The article of claim 7, wherein said at least one element is present at a concentration of up to about 3 atomic percent.
 - 9. The article of claim 2, wherein said coating further comprises up to about 0.1 atomic percent of at least one of carbon and boron.
 - 10. The article of claim 1, wherein said coating has a thickness of greater than about 10 micrometers.
- 10 11. The article of claim 10, wherein said thickness is in the range from about 10 micrometers to about 100 micrometers.
 - 12. The article of claim 11, wherein said thickness is in the range from about 25 micrometers to about 75 micrometers.
- 13. The article of claim 1, wherein said substrate comprises at least one of a nickel-based alloy, a cobalt-based alloy, and an iron-based alloy.
 - 14. The article of claim 13, wherein said substrate comprises a superalloy.
 - 15. The article of claim 14, wherein said substrate comprises a component of a gas turbine assembly.
- 16. The article of claim 15, wherein said component comprises at least one of a turbine airfoil, a turbine disk, and a combustor.
 - 17. The article of claim 1, wherein said single phase of said coating comprises a B2-structured nickel aluminide (NiAl) phase.
 - 18. An article comprising:

a metallic substrate;

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a substantially single-phase B2-structured nickel aluminide coating disposed on said substrate, wherein said coating comprises Ni, Cr, Zr, up to about 20 atomic percent Co, up to about 20 atomic percent Fe, and at least about 30 atomic percent Al, wherein said coating further comprises a gradient in Al composition, said gradient extending from a first Al concentration level at an outer surface of said coating to a second Al concentration level at an interface between said substantially single-phase coating and said substrate;

wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.

10 19. A coating for protecting an article, said coating comprising:

a substantially single-phase coating disposed on a substrate, wherein said coating comprises nickel (Ni) and at least about 30 atomic percent aluminum (Al), wherein said coating further comprises a gradient in Al composition, said gradient extending from a first Al concentration level at an outer surface of said coating to a second Al concentration level at an interface between said substantially single-phase coating and said substrate;

wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.

- 20. The coating of claim 19, further comprises at least one of chromium (Cr), zirconium (Zr), up to about 20 atomic percent cobalt (Co), and up to about 20 atomic percent iron (Fe).
 - 21. The coating of claim 19, wherein said Cr is present at a concentration of up to about 15 atomic percent.
- 22. The coating of claim 21, wherein said Cr is present at a concentration in the range from about 4 atomic percent to about 12 atomic percent.

- 23. The coating of claim 20, wherein said Zr is present at a concentration of up to about 2 atomic percent.
- 24. The coating of claim 23, wherein said Zr is present at a concentration in the range from about 0.2 atomic percent to about 0.8 atomic percent.
- 5 25. The coating of claim 20, wherein said coating further comprises at least one element selected from the group consisting of hafnium (Hf), yttrium (Y), silicon (Si), titanium (Ti), lanthanum (La), cerium (Ce), and tantalum (Ta).
 - 26. The coating of claim 25, wherein said at least one element is present at a concentration of up to about 3 atomic percent.
- 10 27. The coating of claim 20, wherein said coating further comprises up to about 0.1 atomic percent of at least one of carbon and boron.
 - 28. The coating of claim 19, wherein said coating has a thickness of greater than about 10 micrometers.
 - 29. The coating of claim 28, wherein said thickness is in the range from about 10 micrometers to about 100 micrometers.
 - 30. The coating of claim 29, wherein said thickness is in the range from about 25 micrometers to about 75 micrometers.
 - 31. The coating of claim 19, wherein said single phase of said coating comprises a B2-structured nickel aluminide (NiAl) phase.
- 20 32. A coating for protecting an article, said coating comprising:

a substantially single-phase B2-structured nickel aluminide coating disposed on a substrate, wherein said coating comprises Ni, Cr, Zr, up to about 20 atomic percent Co, up to about 20 atomic percent Fe, and at least about 30 atomic percent Al, wherein said coating further comprises a gradient in Al composition, said gradient extending from a

25 first Al concentration level at an outer surface of said coating to a second Al

concentration level at an interface between said substantially single-phase coating and said substrate;

wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.

5 33. A method for protecting an article from a high-temperature oxidative environment, said method comprising:

providing a substrate;

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disposing a first coating layer onto said substrate, wherein said first coating layer comprises nickel (Ni) and aluminum (Al); and

- disposing a second coating layer onto said first coating layer, wherein said second coating layer comprises at least about 90 atomic percent aluminum.
 - 34. The method of claim 33, wherein disposing said second coating layer comprises disposing said second layer using a physical vapor deposition technique selected from the group consisting of electron beam physical vapor deposition and ion plasma deposition.
 - 35. The method of claim 33, wherein disposing said second coating layer comprises disposing a layer comprising at least about 95 atomic percent aluminum.
 - 36. The method of claim 33, wherein disposing said second coating layer comprises disposing a layer consisting essentially of aluminum.
- 20 37. The method of claim 33, wherein disposing said second coating layer comprises disposing a layer having a thickness of at least about 5 micrometers.
 - 38. The method of claim 37, wherein said thickness is in the range from about 5 micrometers to about 20 micrometers.

- 39. The method of claim 33, wherein disposing said first layer comprises disposing said first layer using a technique selected from the group consisting of ion plasma deposition, electron beam physical vapor deposition, thermal spray deposition, and plasma spray deposition.
- 5 40. The method of claim 33, wherein disposing said first layer comprises disposing a first layer further comprising at least one of chromium (Cr), zirconium (Zr), up to about 20 atomic percent cobalt (Co), and up to about 20 atomic percent iron (Fe).
 - 41. The method of claim 40, wherein said Cr is present in said first layer at a concentration of up to about 15 atomic percent.
- 10 42. The method of claim 41, wherein said Cr is present in said first layer at a concentration in the range from about 4 atomic percent to about 12 atomic percent.
 - 43. The method of claim 40, wherein said Zr is present in said first layer at a concentration of up to about 2 atomic percent.
- 44. The method of claim 43, wherein said Zr is present at a concentration in the range from about 0.2 atomic percent to about 0.8 atomic percent.
 - 45. The method of claim 40, wherein disposing said first layer comprises disposing a first layer further comprising at least one element selected from the group consisting of hafnium (Hf), yttrium (Y), silicon (Si), titanium (Ti), lanthanum (La), cerium (Ce), and tantalum (Ta).
- 20 46. The method of claim 33, wherein disposing said first coating layer comprises disposing a layer having a thickness of greater than about 10 micrometers.
 - 47. The method of claim 46, wherein said thickness is in the range from about 10 micrometers to about 100 micrometers.
- 48. The method of claim 47, wherein said thickness is in the range from about 25 micrometers to about 75 micrometers.

- 49. The method of claim 33, further comprising reacting said first coating layer with said second coating layer to form a reacted coating layer.
- 50. The method of claim 49, wherein said reacted coating layer comprises a substantially single-phase coating comprising an aluminide intermetallic compound.
- 5 51. The method of claim 50, wherein said single phase of said reacted coating layer comprises a B2-structured nickel aluminide (NiAl) phase.
 - 52. The method of claim 50, wherein said reacted coating layer further comprises a gradient in Al composition, said gradient extending from a first Al concentration level at an outer surface of said reacted coating layer to a second Al concentration level at an interface between said substantially single-phase coating and said substrate, wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.
 - 53. The method of claim 49, wherein reacting comprises heat-treating said first layer and said second layer.
- 15 54. The method of claim 53, wherein heat-treating comprises heating said substrate in situ during deposition of said second coating layer.
 - 55. The method of claim 33, wherein providing said substrate comprises providing at least one of a nickel-based alloy, a cobalt-based alloy, and an iron-based alloy.
- 56. The method of claim 55, wherein providing said substrate comprises providing a superalloy.
 - 57. The method of claim 56, wherein providing said superalloy comprises providing a component of a gas turbine assembly.
 - 58. The method of claim 57, wherein providing said component comprises providing at least one of a turbine airfoil, a turbine disk, and a combustor.

- 59. The method of claim 33, further comprising disposing a thermal barrier coating over one of said second coating layer and said reacted coating layer.
- 60. The method of claim 59, wherein disposing said thermal barrier coating comprises disposing a ceramic material.
- 5 61. The method of claim 60, wherein disposing said ceramic material comprises disposing a material comprising yttria-stabilized zirconia.
 - 62. A method for protecting an article from a high-temperature, oxidative environment, said method comprising:

providing a metallic substrate;

disposing a first coating layer onto said substrate, wherein said first coating layer comprises nickel (Ni), aluminum (Al), chromium (Cr), zirconium (Zr), up to about 20 atomic percent cobalt (Co), and up to about 20 atomic percent iron (Fe);

disposing a second coating layer onto said first coating layer using a physical vapor deposition technique selected from the group consisting of electron beam physical vapor deposition and ion plasma deposition, wherein said second coating layer consists essentially of aluminum; and

reacting said first coating layer with said second coating layer to form a substantially single-phase reacted coating layer comprising a B2-structured nickel aluminide and further comprising a gradient in Al composition, said gradient extending from a first Al concentration level at an outer surface of said reacted coating layer to a second Al concentration level at an interface between said substantially single-phase coating and said substrate, wherein said first Al concentration level is greater than said second Al concentration level and said second concentration level is at least about 30 atomic percent Al.

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